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氏　　名	野口　　修
授与した学位	博　士
専攻分野の名称	理　学
学位授与番号	博甲第３８７２号
学位授与の日付	平成２１年　３月２５日
学位授与の要件	自然科学研究科　物質分子科学専攻 （学位規則第５条第１項該当）
学位論文の題目	Studies on Trace Elements Analysis by Inductively Coupled Plasma Spectrometry and Its Application to Forensic Chemistry （誘導結合プラズマ分析法を用いた微量元素の分析及び法化学への応用に関する研究）
論文審査委員	教授　本水　昌二　　教授　山本　峻三　　准教授　大島　光子

学位論文内容の要旨

In the field of forensic chemistry, the discrimination of the sample is required in almost all cases. In such cases, it is effective to compare the distribution pattern of multi-elements concentration rather than precise determination of only a target element. Recently, inductively coupled plasma -atomic emission spectrometry (ICP-AES) and inductively coupled plasma -mass spectrometry (ICP-MS) are widely used for analyzing trace of inorganic impurities. ICP-AES and ICP-MS can determine multi-elements simultaneously with high sensitivity. However, volume of the sample solution for the measurement by ICP-AES or ICP-MS is required at least 10 mL. As the volume of forensic samples is usually very small (< 1 mL) and the concentration is trace level, it is very difficult to apply ICP-AES or ICP-MS to forensic samples. Moreover, matrix elements such as Ca, Na and so on, interfere with the determination of trace elements in forensic samples.

The aim of this research is to develop the analytical method for trace heavy metals in forensic samples by using convenient ICP-AES and ICP-MS. Various forensic samples contain heavy metals, and their distribution patterns are characteristic. To achieve this aim, the sample consumption for the measurement must be decreased, and/or matrix elements in samples should be eliminated before measurement.

The analytical method with small sample volume for ICP-MS was developed by air-segmented sample injection (ASSI) and applied to human hair as a forensic sample. The merit of this method is to avoid the dispersion of sample zone by sandwiching with air. The developed method coupled with ICP-MS, could simultaneously determine 25 elements at the sub-ppb (10^{-10} g mL^{-1}) level with only 0.2 mL of a sample solution. This method was successfully applied to a human hair sample, the volume of which was 3 mL (0.015 g of hair).

To develop the more practical measurement method, several on-line collection/concentration systems combined with ICP-AES were examined. Four kinds of chelating resins packed in the mini-column were used for sample pretreatment: collecting the trace elements and eliminating the matrix elements in sample. Among them, laboratory-assembled on-line collection/concentration system which was using commercially available chelating resin, TE-05, was applied to the concrete with about 10 mg of samples. In this case, air-sandwiched method in which the highest elements concentration zone of the eluent was sandwiched by air to improve the sensitivity was developed. The procedure was only increasing the operation steps of the program developed in our laboratory. Major elements (Al, Fe, and Mg) measured by conventional/ICP-AES and trace heavy metals measured by this method, such as Cd, Co, Ni and Pb, were determined without matrices interference. Four concrete samples can be discriminated by comparing the content profiles of the trace elements and the major elements.

As a solid phase for preconcentration, chitosan resin modified with 3-nitro-4-amino benzoic acid (CCTS-NABA resin) and with glycine (CCTS-Gly resin) were newly synthesized and evaluated the performance as sample pretreatment resin. The CCTS-NABA resin could adsorb Mo and CCTS-Gly resin could adsorb Bi almost completely with high selectivity at acidic regions, respectively.

論文審査結果の要旨

In the field of forensic chemistry, the discrimination of the sample is required in almost all cases. In such cases, it is effective to compare the distribution pattern of multi-elements concentration rather than precise determination of only a target element. Recently, inductively coupled plasma -atomic emission spectrometry (ICP-AES) and inductively coupled plasma -mass spectrometry (ICP-MS) are widely used for analyzing trace of inorganic impurities. ICP-AES and ICP-MS can determine multi-elements simultaneously with high sensitivity. As the volume of forensic samples is usually very small (< 1 mL) and the concentration is trace level, it is very difficult to apply ICP-AES or ICP-MS to forensic samples. Moreover, matrix elements such as Ca, Na and so on, interfere with the determination of trace elements in forensic samples.

The aim of this research is to develop the analytical method for trace heavy metals in forensic samples by using convenient ICP-AES and ICP-MS

In this research, the analytical method with small sample volume for ICP-MS was developed by air-segmented sample injection (ASSI) and applied to human hair as a forensic sample. The merit of this method is to avoid the dispersion of sample zone by sandwiching with air. The developed method coupled with ICP-MS, could simultaneously determine 25 elements at the sub-ppb (10^{-10} g mL^{-1}) level with only 0.2 mL of a sample solution. This method was successfully applied to a human hair sample, the volume of which was 3 mL (0.015 g of hair).

Further, to develop the more practical measurement method, several on-line collection/concentration systems combined with ICP-AES were developed. Four kinds of chelating resins packed in the mini-column were used for sample pretreatment: collecting the trace elements and eliminating the matrix elements in sample. A laboratory-assembled on-line collection/concentration system which installed a mini-column packed with commercially available chelating resin, TE-05, was applied to the analysis of concrete with about 10 mg of samples. In this case, air-sandwiched method in which the highest elements concentration zone of the eluent was sandwiched by air to improve the sensitivity was developed. The procedure was only increasing the operation steps of the program developed in our laboratory. Major elements (Al, Fe, and Mg) measured by conventional/ICP-AES and trace heavy metals measured by this method, such as Cd, Co, Ni and Pb, were determined without matrices interference.

As the result of the evaluation of the thesis and the oral presentation, the thesis submitted to the committee was decided to be worthy of a doctoral thesis (Doctor of Philosophy in Science